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Published Version

Peer-review status of attached file:

Peer-reviewed

Citation for published item:

Emziane, M. and Durose, K. and Halliday, D. P. and Bosio, N. and Romeo, N. (2005) 'Efficiency improvement in thin film solar cell devices with oxygen containing absorber layer.', *Applied physics letters*, 87 (26). p. 261901.

Further information on publisher's website:

<http://dx.doi.org/10.1063/1.2152108>

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Efficiency improvement in thin-film solar cell devices with oxygen-containing absorber layer

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(Received 15 July 2005; accepted 27 October 2005; published online 19 December 2005)

The CdTe/CdS solar cell devices were grown using a dry process consisting of sputtering for the transparent conducting oxide and CdS window layers, and close-space sublimation for CdTe absorber layer. These devices were back contacted using Mo/Sb₂Te₃ sputtered layers following the CdCl₂ activation process carried out in air. It was shown that when oxygen is intentionally introduced in the CdTe layer during its growth, this leads to a significant improvement in all the device parameters yielding an efficiency of 14% compared to 11.5% for devices fabricated in the same conditions but without intentional oxygen incorporation in CdTe. The data obtained were not altered following a light soaking. The devices were investigated by quantitative secondary ion mass spectrometry, which allowed insight into the distribution and amount of oxygen and chlorine within the entire device structure. Both impurities showed an increased concentration throughout the CdTe absorber layer. © 2005 American Institute of Physics. [DOI: 10.1063/1.2152108]

A sustained research effort has been made in the past decades towards improving the performance of CdS/CdTe solar cell devices. However, the best efficiencies achieved so far are still about 50% of the theoretically possible performance. Among the approaches used to reach this goal, only a few dealt with the doping of either the CdTe absorber layer or the CdS window layer during their growth.^{1,2} Unfortunately, these attempts led to conflicting results concerning the effect that such a doping could have on the behavior of the resulting device. Indeed, Boyle *et al.* studied CdTe/CdS solar cell devices and concluded that there was neither an obvious correlation between the reagent concentrations in the CdS bath and the solar cell efficiency of CdTe/CdS devices, nor a straightforward relationship between the impurities profiled in the device and the characteristics of the latter.² Similarly, Altosaar *et al.* recently reported on CdS films prepared by chemical bath deposition from solutions containing different donor impurities and found that the performance of solar cells made with the resulting different CdS layers was not noticeably affected.³ This was attributed to the fact that, for these cells, the CdS layer is not necessarily part of the heterojunction with the absorber layer, and that a working buried homojunction exists instead, where the *n*-type region is formed on the surface of the *p*-type absorber.³ However, it was recently shown that the presence of impurities in doped CdS-based thin films leads to an enhancement of the photosensitivity while the photovoltaic efficiency of these films in a metal-semiconductor junction is decreased.¹

In recent works, we reported secondary ion mass spectrometry (SIMS) studies of CdTe/CdS/TCO solar cell structures depth profiled throughout using quantitative SIMS from the back⁴ and the front side.⁵ These investigations demonstrated the existence and importance of impurities in the solar cell structures and elucidated the main sources of impurities. We have also shown that the subsequent overgrowth of

the CdTe absorber layer on top of the CdS window layer substantially affects both the concentration and the profile of the impurity species present in the as-grown CdS (Ref. 6).

In recent reports by different groups, the role of oxygen in CdTe solar cells was mainly investigated for devices post-growth processed in oxygen-containing environments.^{7–9} This investigation aimed at studying the effect, on the characteristics and performance of CdTe/CdS solar cell devices, of an *in situ* doping of the CdTe absorber layer with oxygen. It was found that the introduction of oxygen into the CdTe layer during its growth improves the efficiency of these devices as compared to the devices without the addition of oxygen.

The devices were grown on sapphire substrates in an attempt to avoid the impurity species such as Na, and probably also Si and O, that were shown to be more likely originating from the commonly used glass substrate.^{4,5} These impurities are known to have a doping effect in both CdTe and CdS materials. The sapphire substrates were 2.5 × 2.5 cm² and 0.5 mm thick, and were thoroughly cleaned prior to the structure deposition. The transparent conducting oxide layer was deposited by reactive sputtering at a substrate temperature around 450 °C and consisted of a fluorine-doped indium tin oxide (ITO:F) film (~1.2 μm thick) followed by a tin oxide (SnO₂) film of about 300 nm thickness. Ar+O₂ atmosphere was used for both layers and CHF₃ was added in the case of ITO:F. The CdS:F layer (~80 nm) was also deposited by reactive sputtering using Ar and CHF₃ flows at a substrate temperature of 200 °C and a total pressure of 5 × 10⁻³ mbar. Close-space sublimation (CSS) was used for the growth of the 6–8 μm thick CdTe layer. During the CdTe growth of one of the devices, oxygen was intentionally incorporated *in situ* by letting O₂ gas into the CSS chamber. For the purpose of comparison, the CdTe layer of the other device was grown under the same conditions but without oxygen introduction. Both device structures then underwent a CdCl₂ processing via thermal evaporation of CdCl₂ on top of the structures and air annealing (400 °C for 30 min), fol-

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TABLE I. Solar cell parameters yielded by the devices grown with and without oxygen introduction into the CdTe layer. The solar cell area is 1 cm^2 in both cases.

	$V_{oc}(\text{mV})$	$J_{sc}(\text{mA}/\text{cm}^2)$	Fill factor	Efficiency (%)
Without oxygen in CdTe	780	23	0.64	11.5
With oxygen in CdTe	812	25	0.69	14

lowed by a chemical etching in bromine-methanol solution. Prior to the back contact deposition, the devices were investigated using quantitative SIMS depth profiling (Cameca IMS 4f) with O and Cs primary ion beams. The back contact was finally deposited by sputtering and consisted of a Sb_2Te_3 layer at 300°C and a Mo layer at room temperature. I - V measurements were carried out using an Oriel solar simulator to determine the device parameters under standard conditions, i.e., AM 1.5 and a light power of $100 \text{ mW}/\text{cm}^2$.

Table I recapitulates the parameters of 1 cm^2 solar cell from both devices grown, respectively, with and without oxygen introduction into CdTe. Compared to the device grown without introduction of oxygen into CdTe, all the solar cell parameters, i.e., V_{oc} , J_{sc} , fill factor, and efficiency, are enhanced for the sample with oxygen-containing CdTe layer as a direct consequence of oxygen incorporation. Furthermore, both samples received a light soaking at a temperature of 100°C and 20 suns for 20–30 min, and this did not alter these parameters. Figure 1 shows the quantitative SIMS depth profiles of O and Cl recorded, starting from the CdTe surface through to the TCO layer for both devices grown with and without oxygen incorporation in CdTe. The layer depth scales were calibrated according to the sputter rate in CdTe, and the SIMS data quantified using CdTe relative sensitivity factors as extracted from CdTe implants. Figure 1 also shows that the CdTe/CdS interface is located at a depth of about $8 \mu\text{m}$ from the CdTe free surface. The concentration of O and Cl in the CdS window layer could not be

determined since this thin ($\sim 80 \text{ nm}$) layer was not resolved. The concentration of oxygen in the CdTe layer is about 1 order of magnitude higher (from $\sim 10^{19}$ to $\sim 10^{20} \text{ cm}^{-3}$) especially from the middle of the layer through to the CdTe/CdS interface, when it is intentionally *in situ* introduced compared to the device without oxygen incorporation. The near surface region of the CdTe layer exhibits a concentration of O around $5 \times 10^{19} \text{ cm}^{-3}$. Chlorine, however, is found to have a concentration in CdTe on average 2–5 times higher when oxygen is introduced into the CdTe layer during its growth. As for the distribution of these two elements within the CdTe layer, O and Cl show rather flat profiles for the sample without O incorporation while, for the sample with O incorporation, they exhibit nonuniform distributions particularly for Cl. This indicates that the *in situ* doping of the CdTe layer with O is not only efficient but also favors the incorporation of Cl into the CdTe layer following the CdCl_2 activation process. We believe that oxygen incorporation in CdTe and the enhanced concentration of Cl in CdTe related to the presence of oxygen may both contribute to the improvement of the solar device parameters. In other words, at least in part, O and consequently Cl are not neutral in CdTe but seem to be electrically active and/or form electrically active complexes within the CdTe absorber layer, leading ultimately to an enhanced overall performance of the CdTe/CdS solar cell. This seems to be the case despite the well-known compensation effect in CdTe. Although O is isoelectronic, its role in CdTe has been a subject of considerable discussion.^{7–10} Cl can be incorporated as either a donor, single substitutional impurity, or as an acceptor, the Cl A-center. The mechanism through which the device improvement occurs deserves further investigation.

In summary, we have shown that an intentional *in situ* introduction of O in the CdTe absorber layer of the CdTe/CdS solar cell device leads not only to an order of magnitude increase in the amount of O but also to an enhanced Cl concentration in CdTe as a consequence of O presence. This increase in the concentration of both O and Cl in CdTe was shown to affect directly the resulting device by improving all its parameters, and leading to an efficiency of 14% compared to 11.5% when O is not intentionally incorporated in CdTe during its growth.

The authors are thankful to the EPSRC (UK) for financial support through Grant No. GR/R39283/01.

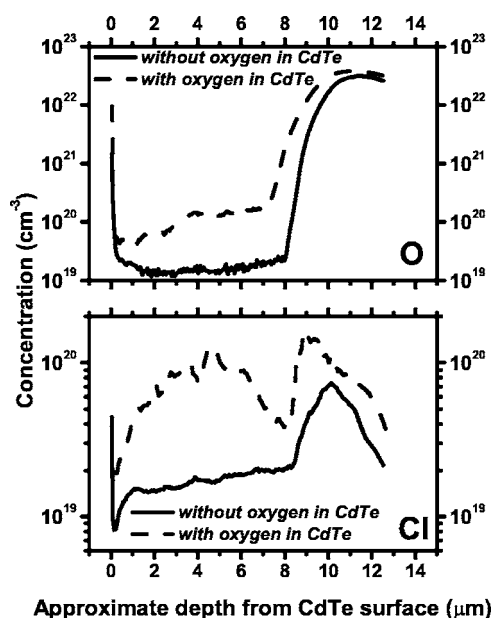


FIG. 1. Quantitative SIMS depth profiles of Cl and O in CdTe/CdS/SnO₂/ITO:F solar cell devices grown with and without intentional incorporation of oxygen in the CdTe absorber layer during its deposition. CdTe relative sensitivity factors were used for the quantification.

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